# Resonant Magnetic Scattering Study of Iron Oxide Layers on Alumina

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## INTRODUCTION

Magnetic oxides, particularly in the form of thin films, are of considerable technological as well as fundamental interest and are the subject of intense research [1]. The epitaxial growth of iron oxides on single crystal oxide substrates leads to the preparation of thin films of high quality. The use of magnetic iron oxides in multilayers opens up new possibilities for the development of spin-polarized tunneling devices [2]. They can also be used as a reference for the investigation of the electronic and magnetic properties of these systems, covering the thicknesses from a single atomic layer to bulk [1,3,4]. Here we describe a resonant scattering experiment of polarized soft x-rays applied to the study of iron oxide epitaxial layers ($\alpha$-$Fe_2O_3$ and $Fe_3O_4$) grown on $\alpha$-$Al_2O_3$. This technique combines the advantages of absorption spectrosocpy (this is the *resonant* aspect) with the structural sensitivity of X-ray scattering. Ferromagnetic ordering can be studied in resonant scattering using either linearly or circularly polarized light on the same footing [5,6], and spectroscopic details can be enhanced selectively by the interplay of the real and imaginary parts of the optical index at resonance [6]. Combined spectroscopic and microstructural studies have been performed on metallic multilayers [7,8], and used to determine the magnetization dependent optical constants [8]. The photon-only aspect of the scattering process is a major advantage for studying insulating samples. We take full advantage of this in our study of magnetic oxide layers in the presence of a magnetic field.

## EXPERIMENTAL

$Fe_3O_4$ (111) and $\alpha$-$Fe_2O_3$ (0001) films were prepared by molecular beam epitaxy assisted by an atomic oxygen source, using alumina (0001) single crystals as substrates. The growth procedure and structural characterization have been described elsewhere [3,9].
Measurements were performed at the *Soft X-ray Metrology* beamline 6.3.2 [10] of the Advanced Light Source storage ring in Berkeley, using the reflectometer endstation. A 70% circular polarisation rate was obtained by asymmetrically positioning the jaws which set the accepted beam divergence. The resolving power at the Fe 2p edges (700-730 eV) was set to 2000. The external magnetic field (about 1 kG) was applied along the intersection between the sample surface and the scattering plane using a permant magnet. The direction of the field was reversed at each acquisition point by rotating the magnet via a computer controlled in-vacuum stepper motor.

## RESULTS AND DISCUSSION

Fig. 1 shows a series of reflectivity curves measured on an 80 Å thick $Fe_3O_4$ film grown on $Al_2O_3$(0001), for several photon energies around the iron 2p edges. The magnetization averaged intensity is reported in the bottom panel, on a semilogarithmic scale. The oscillations of the intensity as a function of $\theta$ come from the interference between the beams reflected at the vacuum / $Fe_3O_4$ and $Fe_3O_4$ / $Al_2O_3$ interfaces, and indicate a total thickness of the film of 85 Å, in

excellent agreement with RHEED and RBS results [11]. The corresponding magnetic signal is reported in the top panel, expressed as the asymmetry ratio $(I^+ - I^-) / (I^+ + I^-)$ between the scattered intensities for the two opposite magnetization/helicity orientations. The different behaviour of the asymmetry ratio curves for different photon energies is related to the energy dependence of the dichroism (see, e.g., Fig. 2) : when going from 709.5 eV to 710.5 eV, for example, the sign of the magnetic signal is reversed, leading to mirror image asymmetry curves in the reflectivity (Fig. 1, top panel).

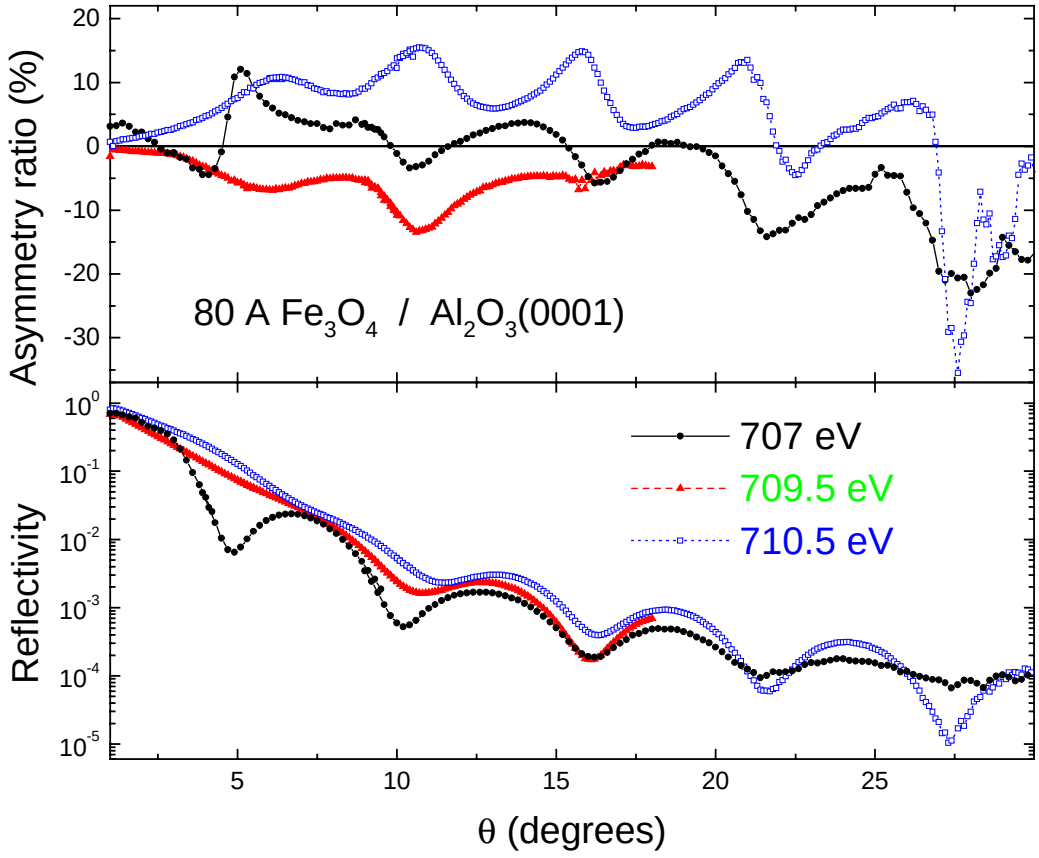


Figure 1. θ/2θ reflectivity scans at three photon energies around the Fe $L_3$ edge for an $Fe_2O_3$ thin film. Bottom: magnetization averaged scattered intensity. Top: asymmetry ratio curves, defined as the difference divided by the sum of the scattered intensity for opposite magnetization / helicity orientations.

We have also investigated thin α-$Fe_2O_3$ films on $Al_2O_3$(0001), using both linearly and circularly polarized light. Strucutral analysis of these samples indicates the formation of a non standard Fe oxide phase during the growth of the first 15-20 Å. Various observations (RHEED, LEED, XPS) suggest that this phase has a 14% lattice expansion with respect to alumina, has an fcc structure like FeO, but contains $Fe^{3+}$ ions [11]. For films thicker than 20 Å only the α-$Fe_2O_3$ phase, of high crystalline quality, is detected. α-$Fe_2O_3$ is an antiferromagnet, so we do not expect to observe magnetic circular dichroism. We have tested this point on an α-$Fe_2O_3$(0001) single crystal: no magnetic signal was detected above a noise level of $10^{-4}$. On the contrary, reflectivity

measurements on an 80 Å $\alpha$-$Fe_2O_3$ / $Al_2O_3$(0001) film indicate a weak (1%) but measurable magnetization dependence.
Fig. 2 compares the energy dependent reflectivity at $\theta$=10° for two 80 Å thick oxide layers, one of composition $Fe_3O_4$ (top panel), the other $Fe_2O_3$ (bottom panel). The magnetic signal is much weaker for $Fe_2O_3$ (note the multiplication factor of 20 on the difference curve), but its shape agrees very well with the one measured for $Fe_3O_4$.

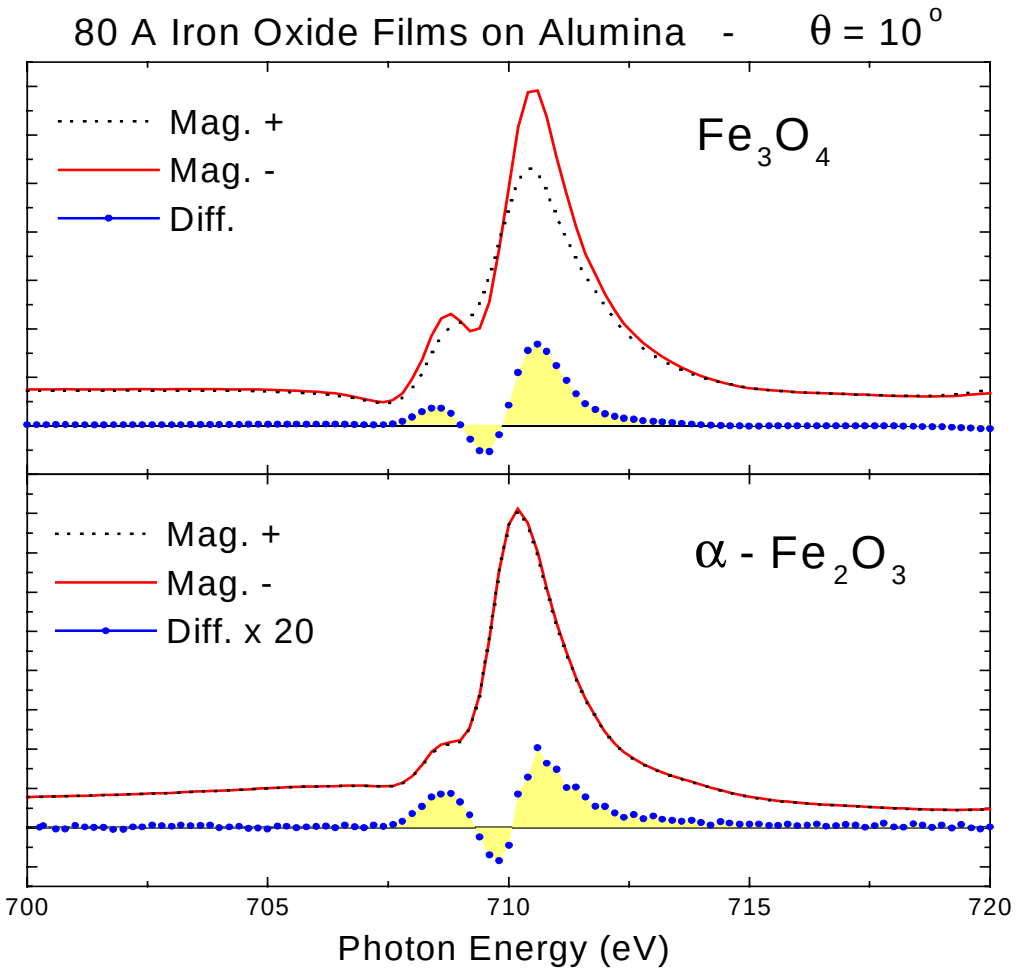


Figure 2. Comparison between $\theta = 10^{\circ}$ reflectivity curves for $Fe_3O_4$ (top) and $Fe_2O_3$ (bottom) films on $Al_2O_3$.

Our data indicate that :

i) the non standard phase observed at the early stage of $\alpha$-$Fe_2O_3$ growth on alumina is not transitory, but remains in the final sample as a sort of naturally formed buffer layer
ii) iron ions contained in this buffer layer carry a net average magnetic moment.

The similarity between dichroism spectra shown in Fig.2 suggests an analogy between the magnetic behaviour of $Fe_3O_4$ and that of the buffer layer, but we cannot conclude, on the basis of this observation only, that $Fe_3O_4$ is present in the $\alpha$-$Fe_2O_3$ thin films, especially because the 2+ valence of iron has never been detected by in situ XPS on thinner films (between 4 and 80 Å). The buffer layer might be $\gamma$-$Fe_2O_3$ like, a ferrimagnetic phase that exhibits nearly the same

inverse spinel structure as $Fe_3O_4$ but contains only ferric ions. Simulations based on atomic calculations including magnetic and crystal field parameters [12] are in progress, in order to determine the local iron environment giving rise to the observed magnetic signals.

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